DETECTION OF CONCEALED MERCURY WITH THERMAL NEUTRONS

Zane W. Bell
Computing and Telecommunications Services Division
Oak Ridge Y-12 Plant*
Martin Marietta Energy Systems, Inc.
Oak Ridge, TN 37381-8142

INTRODUCTION

In the United States today, governments at all levels and the citizenry are paying increasing attention to the effects, both real and hypothetical, of industrial activity on the environment. Responsible modern industries, reflecting this heightened public and regulatory awareness, are either substituting benign materials for hazardous ones, or using hazardous materials only under carefully controlled conditions. In addition, present-day environmental consciousness dictates that we deal responsibly with legacy wastes.

The decontamination and decommissioning (D&D) of facilities at which mercury was used or processed presents a variety of challenges. Elemental mercury is a liquid at room temperature and readily evaporates in air. In large mercury-laden buildings, droplets may evaporate from one area only to recondense in other cooler areas. The rate of evaporation is a function of humidity and temperature; consequently, different parts of a building may be sources or sinks of mercury at different times of the day or even the year. Additionally, although mercury oxidizes in air, the oxides decompose upon heating. Hence, oxides contained within pipes or equipment, may be decomposed when those pipes and equipment are cut with saws or torches. Furthermore, mercury seeps through the pores and cracks in concrete blocks and pads, and collects as puddles and blobs in void spaces within and under them.

Mercury is a hazardous to the health of humans and wildlife. It can be ingested through the food chain (fish, for example, accumulate mercury in their fatty tissue), inhaled as a vapor, or absorbed directly through the skin. Acute inhalation may result in a cough, pneumonitis, pulmonary edema and hemorrhage, fever, vomiting and diarrhea, headache, fatigue, irritability, and ulceration of the mouth and lips. In humans chronic exposure to the vapor affects the kidneys causing kidney disease, the eyes causing discoloration of the lens and opacity of the cornea, the nervous system causing tremors,

*Managed by Martin Marietta Energy Systems, Inc., for the U.S. Department of Energy under contract DE-AC05-84OR21400.

memory loss, and psychological disorders, and the reproductive system. Mercury crosses the placenta and can affect fetal health. The well-known expression "mad as a hatter" derives from the psychological effects observed among workers in the fur hat industry induced by prolonged exposure to mercury. During the 18^{th} and 19^{th} centuries, it was the practice to wash fur hats in mercury to remove oils from the pelts.

Since chronic exposure to mercury can lead to such unpleasant consequences, it is necessary to locate pockets of the material and remove them in a controlled fashion. That is, it is necessary from both the regulatory and ethical points of view to perform the D&D of a facility in such a way as to minimize releases of mercury to the ground, water, and the air. While this task is relatively straightforward when mercury is in pipes or tanks that can be drained, or in equipment that can be dismantled, or even in puddles on the floor (these can be removed with a vacuum cleaner), mercury that has collected inside hollow spaces either as a liquid, oxide, or amalgam presents difficulties. Foremost among them is the problem of detection and quantification. Various technologies for these are described below.

Consider mercury concealed within a hollow wall. Often, there is convenient access to only one side of the wall. Hence, a DXT probe using a low energy x or gamma ray may not be appropriate. (Such a probe is most appropriate for determining the level of elemental mercury in pipes or tanks.) A gauge based on Compton backscattering might be useful only if the wall is sufficiently thin so as not to absorb significant amounts of radiation. Similarly, a device based on x ray resonance fluorescence would be hindered by the absorption of both the excitation x rays and the fluorescent x rays. If the wall is painted, a vapor detector cannot "sniff" out the mercury because layers of paint effectively block the vapor. In scenarios in which walls are not painted, there could conceivably be sufficient ambient vapor to make unambiguous location of a pocket impossible. If the mercury is chemically combined (as in an oxide or amalgam), then devices based on obtaining chemical signatures without contact are also infeasible.

Thus it is seen that the inherent problems encountered in the detection and quantification of concealed mercury are related to the penetration of the probe to the mercury and the escape of the signature to the detector. Neutrons are an attractive probe because they interact only with the nuclei of atoms, and do not distinguish between elemental and chemically combined species. They behave essentially as an inert gas at low pressure and have ranges in concrete and steel that varies from centimeters to meters, depending on energy. Their interactions with nuclei often result in artificial radioactivity and/or the emission of gamma rays, especially upon capture.

Mercury possesses a thermal neutron capture cross section that is two orders of magnitude larger than that of most materials. Upon capturing a neutron, an energetic gamma ray, which can penetrate centimeters of concrete or steel, is emitted. The penetrating power of probe neutrons and gamma ray signature has made it worthwhile to investigate a detection scheme based on capture gamma rays.

The remainder of this paper discusses the design and development of a gauge to detect mercury, regardless of its chemical state, based on the detection of neutron capture gamma rays. The basic physics is described, the design of the neutron source is detailed, and the considerations for the detector are given. The results of bench-top tests are shown, and the status of the system is given.

BASIC PRINCIPLES

Natural mercury has a thermal neutron capture cross section of 384 barns and occurs naturally as 7 isotopes[1]. Almost all the cross section is due to the 2200 barn cross section of ^{199}Hg which comprises 17% of the natural. The absorption of neutrons as they traverse a layer of mercury is given by $e^{-\sigma\rho t}$ where t is the thickness of the layer, σ is the capture cross section, and ρ is the atom density of the mercury. The large cross section of mercury means that a 3 mm thick layer of natural mercury captures 99% of the thermal neutrons incident on it via the reaction $^{199}Hg(n,\gamma)^{200}Hg$.

Capture of a neutron produces ^{200}Hg in an excited state. In 77% of captures, the deexcitation proceeds through the first excited state of ^{200}Hg and results in the emission of a gamma ray with energy 368 keV[2]. This gamma ray can penetrate centimeters of concrete or steel and is therefore a detectable signature of the presence of mercury.

For a flux of neutrons, ϕ, irradiating a mass of mercury, M, the rate, r, of captures is given by

$$r = \frac{\phi \cdot \sigma \cdot M \cdot N_A}{W} \tag{1}$$

where N_A is Avogadro's number, and W is the atomic weight of mercury. The rate r is inferred from the count rate of the 368 keV capture gamma ray corrected for the 77% branching ratio and absorption in the material matrix. Although inversion of equation 1 to obtain M is obviously trivial if r is measured and ϕ is known, this simple treatment is seldom possible because the estimates of r and ϕ are flawed. The estimate of r is uncertain because of assumptions made about the material through which the gamma rays must pass, while that of ϕ is uncertain because of scattering of neutrons by the matrix. In addition, since the capture cross section of mercury is so high, there is significant self-absorption of the neutron flux and the entire mass of a large blob is not exposed to the neutrons. For these reasons, the work to date has concentrated on the development of a mercury detecting device.

In order to get capture gamma rays from mercury, it is necessary to get neutrons to the mercury. In the present work, a radioactive Am-Li source was used to generate neutrons. This source produces a spectrum with energies up to about 1.5 MeV and an average energy of about 340 keV. Since thermal neutrons are required (the capture cross section of mercury falls like $1/\sqrt{E}$ until about 5 eV, and then averages about 15 barns with many resonances at higher energies[3]), it is necessary to degrade the energies of the neutrons by surrounding the source with a moderator. In addition, it is necessary to direct neutrons toward the volume under interrogation. To accomplish these goals, the moderator was designed with the simulation program, MCNP.

MCNP is a Monte Carlo code developed by Los Alamos National Laboratory for the design of neutron shielding and criticality calculations. There have been over 300 person-years invested in its development and it is used over 500 times per month at Los Alamos alone[4]. MCNP determines the behavior of neutrons in the presence of materials by analytically modelling the geometry and then tracking many thousands of simulated neutrons through the material matrix. At each step interactions are selected

according to the relative probabilities of each occurring (that is, the probability of a particular interaction is proportional to the cross section for that process). By simulating the passage of hundreds of thousands of neutrons, estimates of the distribution of neutron energies, direction of travel, probability of capture, and the like may be obtained.

Figure 1 shows a cross section of the final design of the moderator. The central structure is a plexiglas cradle holding a cylindrical source. The cradle/source is surrounded by a polyethylene reflector carved in the shape of an inverted letter W. The opening at the bottom is approximately 36 cm square. The cradle is held in place from below by two plexiglas T bars. This design exposes maximum reflector area to the volume under interrogation (presumably below the moderator) and minimizes the number of neutrons leaking through the reflector. MCNP reports that approximately 40% of the source neutrons are directed toward the target volume, and thermal neutrons make up 80% of those.

Mercury, of course, is not alone in its ability to capture neutrons and emit gamma rays. Notable among other common elements is iron, the main isotope of which, ^{56}Fe, emits gamma rays at 352.4 and 366.8 keV via the $^{56}Fe(n,\gamma)^{57}Fe$ reaction. These gamma rays occur in 10.9% and 1.5% of neutron captures, respectively. In addition, a natural source, 351.9 keV from ^{214}Pb, from the decay chain of uranium, is often found in lead used for the shielding of detectors. The proximity of these gamma ray energies to that from mercury means that a high purity germanium (HPGe) detector must be used to distinguish them from each other. Such detectors have approximately 1 keV energy resolution at 370 keV in contrast to NaI which may have 40 keV resolution. An HPGe detector was used for the present work.

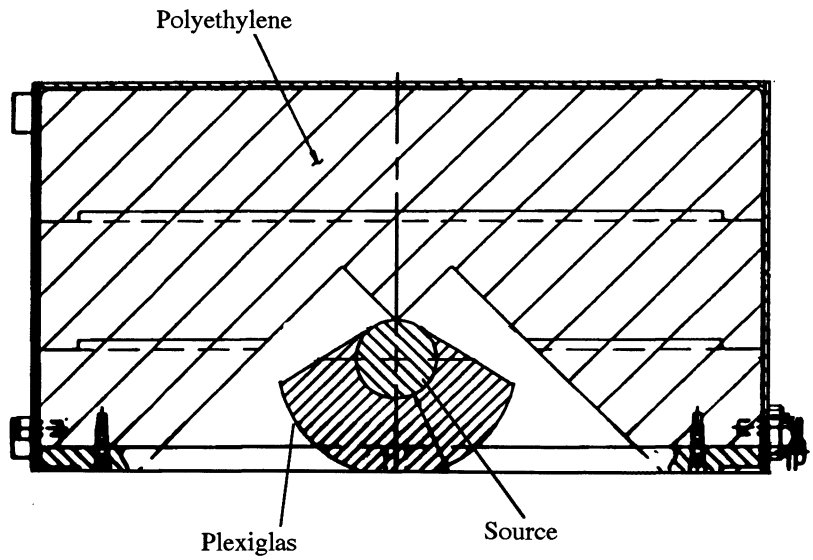

Figure 1. Cross section of moderator/source cradle.

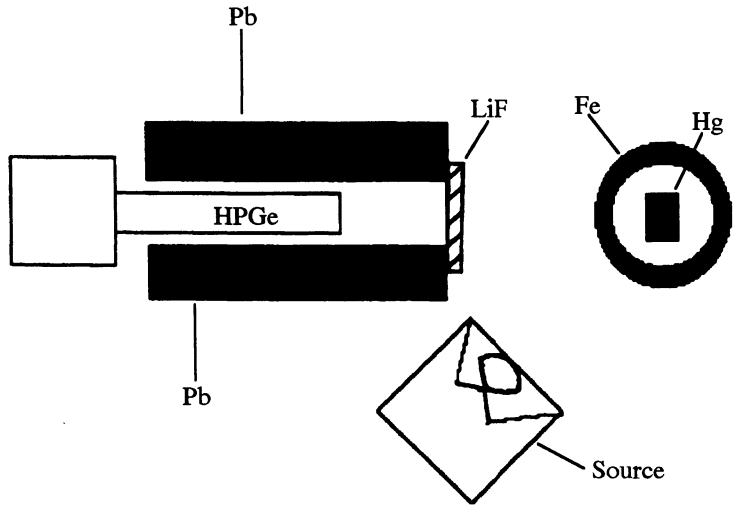

Figure 2. Schematic arrangement of experiment (not to scale).

Figure 2 shows schematically the arrangement of the source, detector and target. The detector is surrounded by a lead collimator wrapped in Cd (to reduce neutron captures in the germanium crystal) and a ^{6}LiF disc. The disc also captures all thermal neutrons incident on it, but emit no capture gamma rays. Thus it does not contribute to the background. The lead collimator is an annulus 9 cm internal diameter, 10 cm thick walls.

Before beginning this work, it was necessary to determine if the neutron source would activate, to any significant extent, structural materials found in buildings. MCNP was used to estimate the behavior of concrete, cold rolled steel, stainless steel, aluminum, copper, and nickel after 10 minutes of irradiation. It was found that those materials whose activation products have the shortest half-lives (Al, Cu, and Na) activate the most. However, they present no danger since they also decay away in a matter of hours. The remaining materials either have very low capture cross sections or half-lives long compared to 10 minutes, and never build up significant amounts of radioactive material. They, too, are of no concern.

RESULTS

The experimental arrangement shown in Figure 2 was used in the laboratory with a 50,000 neutron/sec Am-Li source. (A gauge for use at a sparsely populated remediation site would use a 500,000 neutron/sec source so that the counting time could be decreased without risk of exposure to other workers.) Five mercury batteries (Mallory TR289) containing 37 grams of mercury each[5] (185 grams in total) were placed inside a 3 kg steel pot about 30 cm from both the source and the detector.

Figure 3 shows a typical spectrum obtained after 800 seconds of counting. The leftmost peak is caused by neutrons captured by Ge in the detector crystal itself. It is useful as a flux monitor. The middle peak is caused by the 352.4 keV iron capture gamma ray and the 351.9 keV gamma ray from ^{214}Pb present in the shielding material. The rightmost peak results from mercury capture gamma rays at 368.1 keV. The

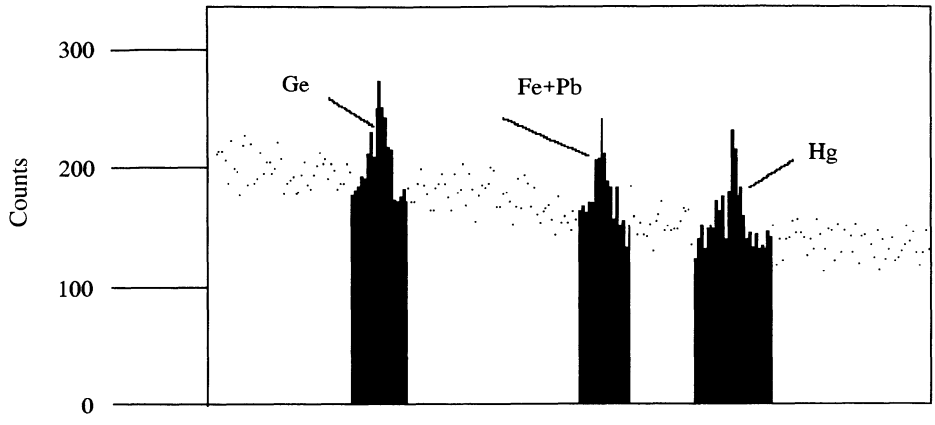

Figure 3. HPGe spectrum showing peaks due to Ge, Fe+^{214}Pb, and Hg.

background on which all these peaks sit is caused primarily by the registration of gamma rays that undergo Compton scattering in the HPGe detector and then escape the Ge crystal.

Table I summarizes the data from spectra (all 800 second counts) taken with various target materials. It obvious that the major component in the Fe+Pb peak in figure 3 is the contribution from ^{214}Pb. From the table, the net counts in the Fe+Pb peak due to iron are 88 ± 74.

The mercury counts are clearly affected by the presence of the steel pot. The linear attenuation for 368 keV gamma rays in steel is approximately 1.3 cm[6]. This means that the 0.25 inch thick walls of the pot account for a loss of 40% of the mercury gamma rays by absorption. This measured values of the mercury counts in rows two and four of the table are consistent with this.

To obtain the true number of mercury gamma rays, it is necessary to correct the mercury counts in the last row of the table for the attenuation in the steel (a factor of 1.63), and for the contribution of the 366 keV iron gamma rays. This correction is 13.7% (1.5/10.9, the ratio of the occurrences of each gamma ray per captured neutron) of the counts due to the 352 keV gamma from iron.

Table I. Contributions to gamma ray spectrum.

Run	Mercury counts	Fe + Pb counts
Background	0	261 ± 27
Mercury only	678 ± 125	263 ± 64
Steel pot only	0	349 ± 69
Mercury and steel pot	435 ± 110	342 ± 73

An estimate of the minimum level of detection of mercury in the presence of iron may be obtained by setting the count rate of the 366 keV iron gamma ray equal to that of the mercury gamma ray. Correcting equation 1 for the numbers of each gamma ray per captured neutron results in the expression shown in equation 2.

$$\frac{M_{Hg}}{M_{Fe}} = \frac{\sigma_{Fe}}{\sigma_{Hg}} \cdot \frac{W_{Hg}}{W_{Fe}} \cdot \frac{0.015}{0.77} \qquad (2)$$

Substitution of the cross sections, and atomic weights yields a ratio of 460ppm by weight. This result is only an statement of the ability of such a gauge to distinguish events due to iron from those due to mercury, and does not include the effects of statistics.

SUMMARY

A scheme based on the detection of thermal neutron capture gamma rays from mercury has been designed and tested in the laboratory. A neutron source and moderator that directs 40% of the source neutrons toward the test volume has also been designed. It has been shown that mercury can be detected even when it is enclosed in 0.25 inch thick steel. It was found that a high purity germanium detector was necessary for this device because of the close (in energy) proximity of capture gamma rays from iron. It is estimated that mercury in the presence of iron may be detected at the 500 ppm level.

A device as described above was originally envisioned as a transportable gauge to be used inside buildings to detect pockets of mercury concealed within hollow walls and pipes. It may be possible to adapt the method to scan a waste stream for mercury and to act a quick sorter. In this mode, the device would be configured similarly to an airport baggage scanner, with material loaded into an irradiation/counting chamber. The device would then need to be fitted either with several HPGe detectors (a very expensive proposition) or with tin or lead loaded plastic scintillators. Such scintillators, although having very poor energy resolution, could be calibrated against the spectra obtained from a single HPGe detector also viewing the counting chamber. Alternatively, the samples to be checked could be put on a turntable and simply scanned. Further investigation along these lines are planned.

REFERENCES

1. F. William Walker, Dudley G. Miller, and Dr. Frank Feiner, *Chart of the Nuclides, 13th Edition*, (General Electric Company, 1983).
2. M. Schmorak, Nuclear Data Sheets 26, 81 (1979).
3. D. I. Garber and R. R. Kinsey, *Neutron Cross Sections BNL 325, Third Edition*, (National Neutron Cross Section Center, Brookhaven National Laboratory, 1976).
4. J. F. Briesmeister, *MCNP 4A CCC-200*, (Radiation Shielding Information Center, Oak Ridge National Laboratory, 1993).
5. Duracell USA, Inc., Material Safety Data Sheet, Sept. 24, 1992.
6. E. F. Plechaty, D. E. Cullen, and R. J. Howerton, *Tables and Graphs of Photon-Interaction Cross Sections From 0.1 keV to 100 MeV Derived from the LLL Evaluated-Nuclear-Data Library* (Lawrence Livermore Laboratory, Livermore CA, 1981).